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FILING DATE.**

**APPLICATION NUMBER: 60/367,231****FILING DATE: March 25, 2002****RELATED PCT APPLICATION NUMBER: PCT/US03/09089**

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COMMISSIONER OF PATENTS AND TRADEMARKS**

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# **PROVISIONAL APPLICATION FOR PATENT COVER SHEET**

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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<input type="checkbox"/> Additional inventors are being named on the _____ separately numbered sheets attached hereto					
TITLE OF THE INVENTION (280 characters max)					
METHOD FOR ABATEMENT OF VOC IN EXHAUST GASES BY WET PULSE CORONA DISCHARGE					
Direct all correspondence to: CORRESPONDENCE ADDRESS					
<input type="checkbox"/> Customer Number		<input style="width: 150px;" type="text"/>		Place Customer Number Bar Code Label here	
OR Type Customer Number here					
<input checked="" type="checkbox"/> Firm or Individual Name		Welsh & Katz, Ltd.			
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ENCLOSED APPLICATION PARTS (check all that apply)					
<input checked="" type="checkbox"/>	Specification Number of Pages	<input style="width: 50px; text-align: center;" type="text" value="11"/>	<input type="checkbox"/> CD(s), Number <input style="width: 100px;" type="text"/>		
<input checked="" type="checkbox"/>	Drawing(s) Number of Sheets	<input style="width: 50px; text-align: center;" type="text" value="4"/>	<input type="checkbox"/> Other (specify) <input style="width: 150px;" type="text"/>		
<input checked="" type="checkbox"/>	Application Data Sheet. See 37 CFR 1.76				
METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT (check one)					
<input checked="" type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27.				FILING FEE	
<input checked="" type="checkbox"/> A check or money order is enclosed to cover the filing fees				AMOUNT (\$)	
<input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number		<input style="width: 100px; text-align: center;" type="text" value="23-0920"/>		<input style="width: 100px; text-align: center;" type="text" value="\$80.00"/>	
<input type="checkbox"/> Payment by credit card. Form PTO-2038 is attached.					
The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.					
<input type="checkbox"/> No.					
<input checked="" type="checkbox"/> Yes, the name of the U.S. Government agency and the Government contract number are: <u>DE-FC07-00ID13868</u>					

Respectfully submitted,

SIGNATURE



Date

03/25/2002

TYPED or PRINTED NAME

Edward P. Gamson

REGISTRATION NO.

29,381

(if appropriate)

Docket Number:

UIK-101.0

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## **USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT**

This collection of information is required by 37 CFR 1.51. The information is used by the public to file (and by the PTO to process) a provisional application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 8 hours to complete, including gathering, preparing, and submitting the complete provisional application to the PTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, Washington, D.C. 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Box Provisional Application, Assistant Commissioner for Patents, Washington, D.C.

P19SMALL/REV05

# FEE TRANSMITTAL for FY 2002

Patent fees are subject to annual revision.

☒ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT **\$80.00**

## Complete if Known

Application Number	Not Assigned
Filing Date	March 25, 2002
First Named Inventor	Alexander Gutsol
Examiner Name	Not Assigned
Group Art Unit	Not Assigned
Attorney Docket No.	UIK-101.0 (8525/87012)

## METHOD OF PAYMENT (check all that apply)

☒ Check ☐ Credit card ☐ Money Order ☐ Other ☐ None

☐ Deposit Account

Deposit Account Number **23-0920**

Deposit Account Name **Welsh & Katz, Ltd.**

The Commissioner is authorized to: (check all that apply)

☐ Charge fee(s) indicated below ☒ Credit any overpayments

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## FEE CALCULATION

### 1. BASIC FILING FEE

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description	Fee Paid
101 740	201 370	Utility filing fee	
108 330	206 165	Design filing fee	
107 510	207 255	Plant filing fee	
108 740	208 370	Reissue filing fee	
114 180	214 80	Provisional filing fee	80.00
<b>SUBTOTAL (1)</b>			<b>\$80.00</b>

### 2. EXTRA CLAIM FEES FOR UTILITY AND

Total Claims	Extra Claims	Fee from below	Fee Paid
Independent Claims	-20** = 0	X	0.00
Multiple Dependent	-3** = 0	X	0.00

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description	Fee Paid
103 18	203 9	Claims in excess of 20	
102 84	202 42	Independent claims in excess of 3	
104 280	204 140	Multiple dependent claim, if not paid	
109 84	209 42	** Reissue independent claims over original patent	
110 18	210 9	** Reissue claims in excess of 20 and over original patent	
<b>SUBTOTAL (2)</b>			<b>\$0.00</b>

\*\*or number previously paid, if greater, For Reissues, see above

## FEE CALCULATION (continued)

### 3. ADDITIONAL FEES

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description	Fee Paid
105 130	205 65	Surcharge - late filing fee or oath	
127 50	227 25	Surcharge - late provisional filing fee or cover sheet	
139 130	139 130	Non - English specification	
147 2,520	147 2,520	For filing a request for ex parte reexamination	
112 920*	112 920*	Requesting publication of SIR prior to Examiner action	
113 1,840*	113 1,840*	Requesting publication of SIR after Examiner action	
115 110	215 55	Extension for reply within first month	
116 400	216 200	Extension for reply within second month	
117 920	217 480	Extension for reply within third month	
118 1,440	218 720	Extension for reply within fourth month	
128 1,960	228 980	Extension for reply within fifth month	
119 320	219 160	Notice of Appeal	
120 320	220 160	Filing a brief in support of an appeal	
121 280	221 140	Request for oral hearing	
138 1,510	138 1,510	Petition to institute a public use proceeding	
140 110	240 55	Petition to revive - unavoidable	
141 1,280	241 640	Petition to revive - unintentional	
142 1,280	242 640	Utility issue fee (or reissue)	
143 480	243 230	Design issue fee	
144 620	244 310	Plant issue fee	
122 130	122 130	Petitions to the Commissioner	
123 50	123 50	Processing fee under 37 CFR § 1.17(q)	
126 180	126 180	Submission of Information Disclosure Statement	
581 40	581 40	Recording each patent assignment per property (times number of properties)	
146 740	246 370	Filing a submission after final rejection (37 CFR § 1.129(a))	
149 740	249 370	For each additional invention to be examined (37 CFR § 1.129(b))	
179 740	279 370	Request for Continued Examination (RCE)	
169 900	169 900	Request for expedited examination of a design application	

Other fee (specify) \_\_\_\_\_

\*Reduced by Basic Filing Fee Paid

**SUBTOTAL (3)**

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## APPLICATION DATA SHEET

### APPLICATION INFORMATION

*Application number::*

*Filing Date::*

03/25/2002

*Application Type::*

Provisional

*Subject Matter::*

Utility

*Suggested Classification::*

*Suggested Group Art Unit::*

*Title::*

METHOD FOR ABATEMENT OF VOC IN  
EXHAUST GASES BY WET PULSE CORONA  
DISCHARGE

*Attorney Docket Number::*

UIK-101.1 (8525/87011)

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No

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Yes

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No

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## REPRESENTATIVE INFORMATION

Registration Number One::	24,003
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Registration Number Five::	25,060
Registration Number Six::	22,053
Registration Number Seven::	27,466
Registration Number Eight::	29,434
Registration Number Nine::	29,054
Registration Number Ten::	29,381
Registration Number Eleven::	34,044
Registration Number Twelve::	27,600
Registration Number Thirteen::	34,137
Registration Number Fourteen::	39,724
Registration Number Fifteen::	37,963
Registration Number Sixteen::	38,110
Registration Number Seventeen::	39,021
Registration Number Eighteen::	41,217
Registration Number Nineteen::	34,177

60367231-03300

METHOD FOR ABATEMENT OF VOC IN EXHAUST GASES  
BY WET PULSE CORONA DISCHARGE

GOVERNMENTAL SUPPORT

The invention was made in the frame of a DOE sponsored project, Project Identification No. DE-FC07-00ID13868. The government has certain rights in the invention pursuant to that support.

DESCRIPTION

There are numerous methods for the waste gases treatments from volatile organic compounds (VOC) with the help of electrical discharges, and particularly with the help of the pulsed corona discharge [Malik et al., *Chinese J. Chem. Eng.*, 7(4) (1999)]. To remove one large VOC molecule with the help of such methods it is necessary to produce several active radicals like OH. The energy price of one radical production is very high - about 50 eV per radical. As a result, total price of one VOC molecule removal (transformation of VOC molecule into H<sub>2</sub>O and CO<sub>2</sub>) is also very high - about 300 eV. It is possible in this case to satisfy the industry demand to spend not more than about 10 W-Hour/m<sup>3</sup> of the waste gas only if VOC concentration is not higher than about 30 ppm. Real industrial waste streams such as that of the papermaking, metal cleaning and plating, plastics manufacture and the like industries have VOC contamination several times higher, so usual plasma methods for VOC removal are not applicable.

A new method has been developed that combines the corona discharge with the scrubber to

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provide a "wet" corona discharge. [Fridman et al., 199<sup>th</sup> Meeting of the Electrochemical Society, Washington, March 25-29, 2001; Meeting Abstracts, Vol. 2001-1, Abstract No. 196; and Sobacchi et al., 15<sup>th</sup> International Symposium on Plasma Chemistry, Orleans, July 9-13, 2001; Symposium Proceedings, Vol. VII: Poster Contributions, pp. 3135-3140.]

To obtain desirable VOC destruction with low energy consumption it necessary to make proper design of pulsed corona discharge and scrubber. This method is applicable to substantially any existing waste gas stream that emanates from an industrial site such as a metal cleaning and plating, paint manufacturing, plastics manufacturing, petroleum refining and dye-making site that also includes a wastewater treatment facility. The contemplated method is illustrated herein in relation to a papermaking facility.

The target streams for this invention such as HVLC Brownstock Washer Vent Emissions can contain various VOCs in varying concentrations, as are seen in Table I, below.

Table I

Emitted VOC component	HVLC Brownstock Washer Vent Emissions according to the information of NCASI	"The worst case scenario" HVLC Brownstock Washer Vent Emissions suggested by GP
Dimethyl Disulfide	2 ppm	20 ppm
Dimethyl Sulfide	-	1727 ppm
Methanol	83 ppm	2330 ppm
Acetone	3 ppm	-
Terpenes	209 ppm	62 ppm
Process Conditions		
Temperature	103°F	150°F
Relative Humidity	100%	100%

In the simplest case (shown schematically in Fig. 1) the wet corona setup permits for a water film to flow on the internal walls of the reactor. In a larger scalable geometry a water spray can be used instead of water film.

The corona discharge is generated in a wire-into-cylinder coaxial electrode system. The inner, high voltage electrode is a 0.5 mm diameter Inconel<sup>®</sup> wire. The Inconel<sup>®</sup> alloy has been selected for its superior heat and oxidation resistance. The external electrode is made up of a 60 cm long cylindrical glass tube with a 22.2 mm internal diameter, surrounded by a sheet of perforated metal.

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The internal wall of the glass tube is covered by a layer of absorbing material, which forms a uniformly distributed water film in the reactor. The tube is held at the top by a Teflon<sup>®</sup> holder and at the bottom by a stainless steel holder; stainless steel was chosen for the bottom of the reactor to prevent deterioration of the holder itself due to the byproducts expected in the liquid phase, such as sulfuric acid.

The reactor is sealed by means of silicon o-rings. The top holder contains the connections of the lines for the incoming water and the outgoing gas; water is introduced into the reactor through sixteen, equally spaced holes with a diameter of  $1/32$ " ( $\sim 0.8$  mm), to guarantee a uniform injection around the circular tube. It also contains the connection for the internal, high voltage electrode and for the high voltage probe. The bottom holder includes threaded holes for the gas inlet and for the water outlet. To avoid any contact between this metal holder and the high voltage (HV) electrode and to prevent the formation of sparks, the wire is fixed to the holder through a Teflon<sup>®</sup> insert, which completely contains the lower portion of the wire.

Power is supplied to the plasma source by a thyatron-based power supply, with 100 k $\Omega$  internal resistance and no-load voltage values from 0 to 20 kV. Pulses of about 100 ns duration and 10 ns rise time are applied; they are transmitted to the central wire electrode by means of a 50  $\Omega$  high-voltage, coaxial cable. The power of the plasma source is varied from 1 to 20 W by controlling the high voltage pulse amplitude and the pulse frequency, which could

range from 0.01 to 2 kHz. Voltage is measured using a high voltage probe (model P6015A 1000X, Tektronik Inc.), current measurements are performed using a standard current monitor (Current monitor #411, Pearson Inc.).

Correct choice of a scrubber parameters should be based on theoretical estimations or experimental results. The solubility of a gas in water is described by the Henry's law, which states a relation between the concentrations of a gas in the liquid and in the gas phases. Usually, the Henry's law constant  $k_H$  is defined as:

$$k_H = \frac{c_a}{p_g} \quad (1)$$

Here,  $c_a$  is the concentration of a species in the aqueous phase and  $p_g$  is the partial pressure of that species in the gas phase. The commonly used unit for the Henry constant is:

$$\left[ \frac{M}{atm} \right] = \left[ \frac{mol_{aq}/m^3}{atm} \right] \quad (2)$$

The higher the value of the Henry's law constant for a given compound, the higher the solubility of that compound in water. Values of the Henry's law constants for the VOCs of interest are shown in Table II, below. Oxidized compounds such as methanol, acetone and the intermediate byproducts from the oxidation of dimethylsulfide and  $\alpha$ -pinene (that are the main substances from terpenes in HVLC Brownstock Washer Vent Emissions) appear to have a much higher solubility than the non-oxidized species.

Table II

Henry's law constant values in standard conditions

	$k_H^\ominus \left[ \frac{M}{atm} \right]$
Methanol $CH_3OH$	$2.2 \cdot 10^2$
Acetone $CH_3COCH_3$	$3.1 \cdot 10^1$
Dimethyl Sulfide $CH_3SCH_3$	$9 \cdot 10^{-1}$
$\alpha$ -Pinene $C_{10}H_{16}$	$4.9 \cdot 10^{-2}$
Butane $C_4H_{10}$	$1.2 \cdot 10^{-3}$
Sulfur Dioxide $SO_2$	$1.2 \cdot 10^1$

The solubility of a gas in water depends on the temperature. When we refer to standard conditions ( $T^\ominus = 298.15$  K), the Henry's constant is denoted as  $k_H^\ominus$ . Henry's law can be described as a function of temperature as:

$$k_H = k_H^\ominus \exp \left[ \frac{-\Delta_{soln} H}{R} \left( \frac{1}{T} - \frac{1}{T^\ominus} \right) \right] \quad (3)$$

where  $\Delta_{soln} H$  is the enthalpy of solution.

A quasi two-dimensional model of VOCs absorption into water was developed. The model is based on the solution of the species conservation equations for the gas phase and for the liquid phase. It yields to the following expression for the Destruction and Removal Efficiency (DRE):

$$DRE = \frac{(1-\alpha) \cdot e^{-\beta(1-\alpha)}}{1-\alpha \cdot e^{-\beta(1-\alpha)}} \quad (4)$$

where  $\beta$  is the ratio between the convection time and the diffusion time and can be written as:

$$\beta = \frac{4\tau_c}{\tau_d} = \frac{\left(\frac{D}{d^2}\right)}{\left(\frac{V_g}{L}\right)} \quad (5)$$

with:  $D$  = diffusion coefficient in air;  
 $d$  = electrode separation distance;  
 $V_g$  = velocity of the gas;  
 $L$  = length of the wet corona reactor; and

$\alpha$  is a ratio between gas and liquid flow rate that is expressed as:

$$\alpha = \frac{Q_g}{Q_w V_{mol} k_H} \quad (6)$$

with:  $Q_g$  = gas flow rate;  
 $Q_w$  = water flow rate;  
 $V_{mol}$  = gas volume per unit mole.

$D$  and  $k_H$  are the only parameters related to the specified compound whose removal is being analyzed; the other factors describe the geometry of the reactor ( $d$  and  $L$ ) and the experimental conditions ( $V_g$ ,  $Q_g$  and  $Q_w$ ).

With the assumption, which holds true if wet corona design is made appropriate, that the convection time is much longer than the diffusion time, i.e.  $\beta \gg 1$ ,  $\alpha$  becomes the critical parameter: for  $\alpha > 1$ , the DRE is very low, whereas for  $\alpha < 1$  high values of DRE are obtained. Because the gas flow rate is fixed for particular technology stream and  $V_{mol}$  and  $k_H$  are constant in standard conditions, assuming  $\alpha = \alpha_{crit} = 1$ , a critical value for the water

flow rate can be derived as:

$$Q_{w-crit} = \frac{Q_g}{V_{mol} k_H} \quad (7)$$

Only a flow rate higher than this critical value provides a high removal of a given substance by absorption into the water film. This theoretical analysis shows good agreement with the experimental results presented below.

Methanol removal by absorption into water was studied in the wet corona reactor. An initial concentration equal to 200 ppm of methanol was considered. Methanol is the most soluble among the organic compounds of interest, as Table II shows. The value of the critical water flow rate for absorbing methanol was calculated from the model to be  $Q_{w-crit} \sim 0.2$  ml/min, in good agreement with the experimental results shown in Fig. 2. For a fairly low flow rate of the water film ( $\sim 0.9$  ml/min), the DRE is almost complete (about 99.9%), suggesting that an optimal configuration in terms of the geometry of the reactor and of the ratio between gas and liquid flow rate has been obtained for the removal of methanol.

Acetone removal by absorption into water was studied for flow rate of the water film varying from 0 to about 2 ml/min. The initial concentration of acetone in the gas stream was 200 ppm. For acetone, with a 1 SLM gas flow rate, the calculated value of the critical water flow rate is  $Q_{w-crit}$  about 1.3 ml/min, higher than for methanol, given the lower solubility in water; experimental results, presented in Fig. 3, show good agreement with the theoretical calculations. A high removal efficiency (about 99%)

can be reached using a fairly low water flow rate (about 2 ml/min).

Thus, absorption into water of compounds with high Henry's law constants, such as methanol and acetone, appears to be a very feasible solution given the low water flow rate required, which is, by weight, comparable to the gas flow rate or even lower; absorption is economically practical for the removal of these compounds from industrial gas streams when water is readily available (as in the case of the paper industry).

According to our invention all soluble VOC components, like methanol or acetone, are removed from the stream by water, and the only one radical OH produced by the pulsed corona discharge is used for treatment of one molecule of insoluble VOC, like dimethylsulfide.

Molecules of insoluble VOCs are non-polar; this is a reason of their very low solubility. After the interaction of OH radical with the molecule of insoluble VOC, this molecule transforms into a new molecule or radical, which is polar and more soluble. Then this more soluble substance is removed by the water stream of the scrubber. As a result not more than 50 eV per molecule of VOC is spent, and this system is applicable for the waste stream with higher level of contamination (about 200 ppm of insoluble VOC and any reasonable level of soluble VOC).

For example, in this system, one need only spend 10 W-Hour/m<sup>3</sup> for the waste gas treatment to reach 90% DRE for the case of the next VOC composition (Table 3).



Table 3

## Gas composition tested in study

Ingredient	VOC content (ppm)
Dimethyl Sulfide	50
Methanol	200
Acetone	200
$\alpha$ -Pinene	50

During this study, the gas flow rate was 1 SLM, and water flow rate was 1.5 ml/min.

Usually, the radicals being dissolved in the water produce acidic solutions ( $\text{pH} < 7$ ). Additionally, ozone molecules from the corona discharge also dissolve in the scrubber water to make it more acidic. As a result, the corona device is built from the materials that are stable to acidic solution: stainless steel, plastics, glass, carbon materials, and the like.

One basis for the application of the present invention to exhaust gases of paper industry or a similar industry is the presence of a huge amount of wastewater in this industry. As a result, a small amount of wastewater provided from the scrubber does not create a problem; i.e., the scrubber wastewater can be mixed with the existing wastewater and the mix can be treated later according to the existing techniques. If at the particular plant the main wastewater is alkaline ( $\text{pH} > 7$ ), an interaction of the scrubber wastewater with the main wastewater stream results in oxidation of the remaining dissolved VOC into  $\text{H}_2\text{O}$  and  $\text{CO}_2$ ; i.e., in the waste mix water cleaning.

If the particular plant has the wastewater that is of such quality that it can be used in spray systems and does not produce considerable amount of

VOCs during spraying, this wastewater can be used in the corona scrubber.

Currently, the Regenerative Thermal Oxidizers (RTO) are used for VOC treatment in the paper industry [see, Harkness et al., NCASI Technical Bulletin No. 795, September (1999)]. These devices are very good for a high level of contamination, when VOC oxidation provides a substantial amount of energy. Otherwise, a lot of natural gas should be used for RTO operation. Other disadvantages of RTO are the SO<sub>2</sub> emission in the typical case of sulfur-contained VOC and cyclic operation resulting in cracking of the ceramic parts of RTO and high capital and operation expenses. The technology described herein is believed to be economical reasonable in the case of low energy consumption as can be seen from the annualized cost comparison between the Wet Corona and RTO processes shown in Fig. 4.

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FIG. 1

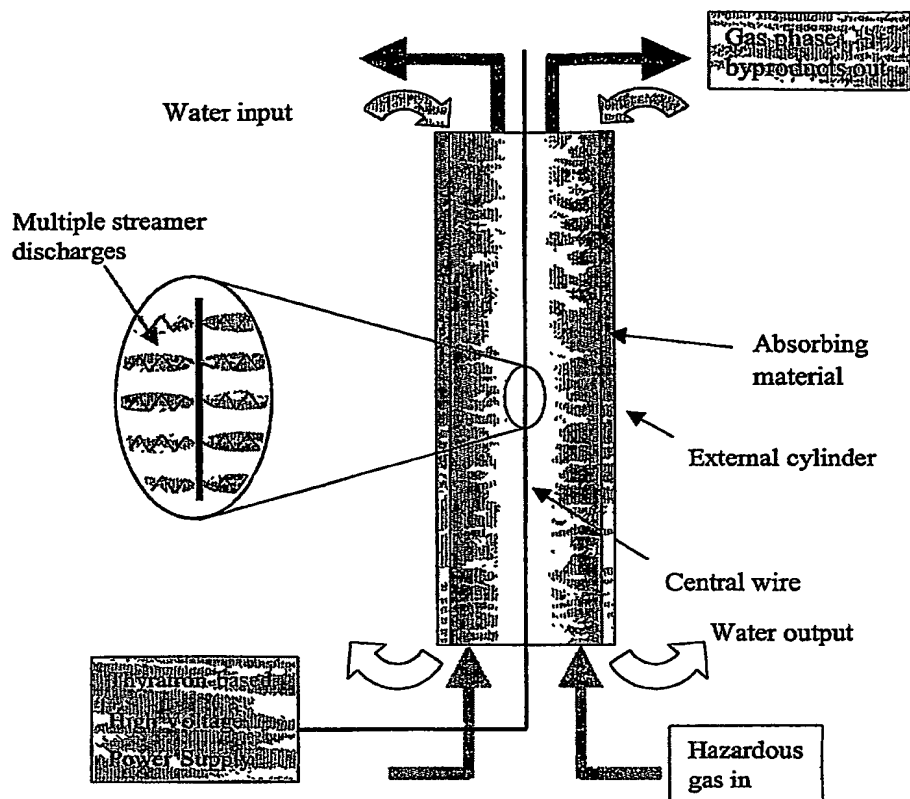
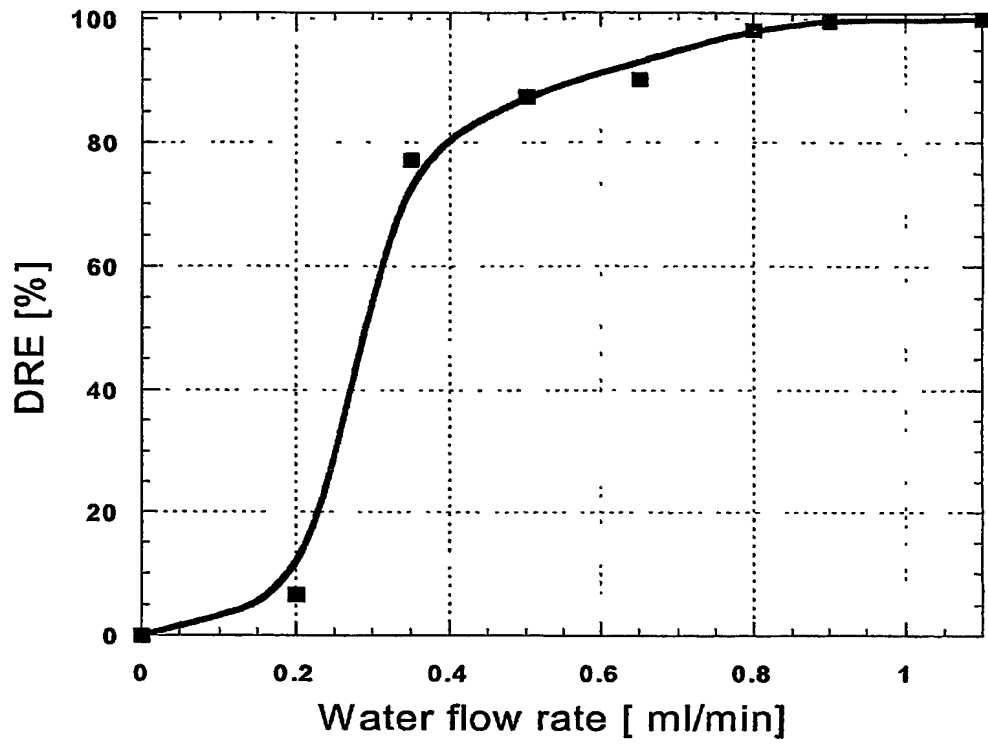


FIG. 2



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Fig. 3

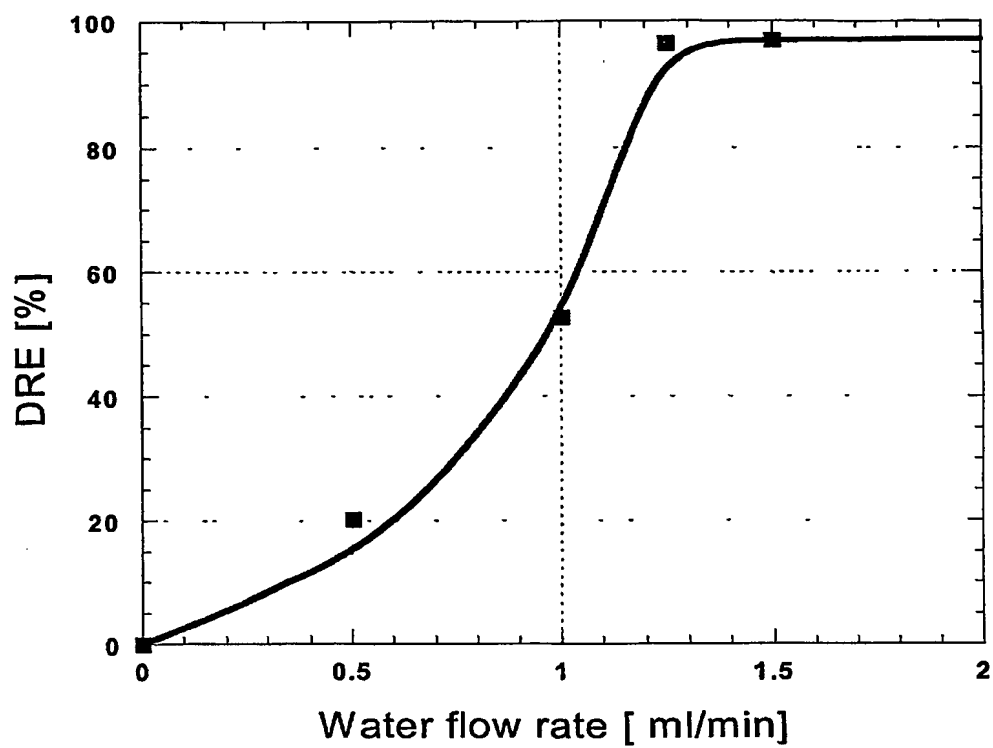


Fig. 4

